Reply to Interactive comments from the Anonymous Referee #2

We would like to thank Anonymous Referee #2 for the helpful and supportive comments. Below we have addressed the comments/questions point-wise. For clarity, we keep the editors comments in blue and our replies are in black font.

General comments

Berhanu et al. report co-located observations of continuous CO2, CO and 14CO2 from grab samples at the Beromünster tower (Switzerland) for 2013-2016. The variability of the mixing ratio gradients relative to the high-alpine research station Jungfraujoch is discussed, especially focussing on the CO2 offset, which is demonstrably affected by both biogenic and fossil fuel CO2 fluxes. Seasonal, episodic and diurnal variations of DCO2,ff and DCO2, bio are interpreted as well as RCO, which is compared to reported emission ratios for Switzerland. The paper is well written, soundly structured and the experimental methods are well described. The tools used to interpret the data are commonly used in this field and the results are presented clearly. Unfortunately, the interpretation of the reported results falls short at several occasions. Although plausible, more care has to be given to substantiate the interpretations.

1.) At several occasions local temperatures are given as likely cause of e.g. large positive DCO2, bio due to high temperatures (L341) or high RCO due to unusually cold conditions (L371). If T is such a strong predictor it should be added to the figures or a regression analysis added to the manuscript. The potential impact of PBL variations on reported mixing ratio gradients is also mentioned, but no thorough analysis is performed.

As the reviewer pointed out, we have ascribed part of the observed strong depletions in radiocarbon or enhanced biospheric fluxes to local temperature. This latter case was also elaborated in a recent study from the same site (Oney et al., 2017). We have now included the temperature record of the site from the highest inlet of the tower (212.5 m), to match with the radiocarbon measurements in Fig. 2e.

We discussed this issue further in reply to reviewer#1 (page 8) and additional information is included in the revised manuscript lines 386-401.

We agree with the reviewer that through analysis of the PBL variations is important to better understand variations in R_{CO} and CO_{2bio}. However, this is beyond the current scope of this study but will be considered in future studies and this information is provided in the conclusion section.

2.) Changes in the area of influence (footprint) are also mentioned as likely causes for specific excursions of the time series, but footprints are not given in the manuscript (or as a supplement).

The simulated footprints from FLEXPART-COSMO are now provided in the supplementary

section.

3.) More broadly, the influence of a (changing) footprint seems to be ignored when the observational RCO is compared to RCO reported in the Swiss emission inventory. Before suggesting that the Swiss emission inventory potentially under-reports CO/CO2 emission ratios, the author need to demonstrate if the observed RCO is representative of average Swiss emissions or how much (and when) RCO is indeed affected by CO ad CO2 emissions from other regions (as mentioned in e.g. L371) plus photochemistry during the trajectory. Overall, the topic is of interest and novel data that could help constrain biogenic CO2 fluxes for (central) Europe can be expected to be of some interest for the ACP readership, if the major comments are addressed.

The Beromünster tower, which is situated on the southern border of the Swiss plateau has been assessed for its foot print in previous publications (Oney et al., 2015; Henne et al., 2016). Both studies show that the measurements are mostly representative of the Swiss plateau (the flat part of Switzerland between the Alps and the Jura mountains where the vast majority of the population lives and where most of the industrial and agricultural activity takes place). The study by Henne et al. (2016) on inverse modelling of CH_4 further revealed an excellent agreement between the top-down estimates and the Swiss methane emissions further suggesting that measurements at the tower capture concentration fluctuations that are representative for emissions in Switzerland.

Specific comments:

L69: The 14C produced in the lower stratosphere is definitely of great interest as it is most easily transported into the troposphere, but 14C is (also/mostly) produced in other altitudes.

Following also the suggestion of the other reviewer, we have changed the term "lower stratosphere" to "upper atmosphere"

L165: How did you ensure that samples taken before the leakage was detected were not (slightly) contaminated? Leak sometimes slowly increase over time before they are noticed or was there an abrupt change in mixing ratios or an identifiable mechanical failure?

Indeed possible minor leaks before detection could not be ignored. However, such minor leaks are expected to manifest themselves in deviations not only in the amount of CO_2 that has been extracted but also in its stable isotope ratio measurements. Yet, not observed such deviations in these periods.

L167: Please correct to "replaced"

We have now corrected the word "replaced"

L203: Please note in the text that equation 2 is only an approximation. For the correct mass-balance for 14CO2 small deltas need to be used (big delta includes an isotopic correction term based on small delta 13C)

We have now modified the paragraph about equation 2 as:

Each of these components has a specific $\Delta^{14}C$ value (i.e. the deviation in per mil of the $^{14}C/^{12}C$ ratio from its primary standard, and corrected for fractionation and decay using ^{13}C measurements) described as $\Delta^{14}C_{meas}$, $\Delta^{14}C_{bg}$, $\Delta^{14}C_{bio}$ and $\Delta^{14}C_{ff}$. In analogy to Eq. (1), a mass balance approximation equation can also be formulated for ^{14}C as…

In case we do not apply this simple correction the CO_{2bio} will be changed by about 0.4 ppm on average.

This is a valid point. Annual total emissions from Beznau were indeed only little smaller in 2015 compared to 2014 despite long maintenance periods. Note that emissions from Beznau were not assumed to be zero after March 2015, because only reactor 1 was shut down, but not reactor 2.

Zelleweger et al. 2012 reported a precision of 2.5 ppb and 1.0 ppb for 1-minute and 10-minute averages, respectively for the CRDS CO-analyzer at JFJ. In the uncertainty calculation we have used the 1-minute uncertainty.

As mentioned above we have now included the FLEXPART-COSMO footprints for each sampling period in the supplementary materials, which is more informative. We believe such modification of Figure 2 will not add additional information but rather complicate the figure, as much information is already included.

Despite the fact that the two RCO values are not significantly different considering the uncertainties, the correlation coefficients are different with r^2 of only 0.3 in summer (Table 1). Hence, a higher uncertainty was calculated for summer, and comparing with the better-constrained winter values, will be misleading. This is also implicated in the paragraphs following this section where we have only considered the wintertime R_{CO}. We have now clarified these points in the manuscript in lines 408-410.

Thank you for your suggestion, we rephrased it in the main text as:

The values obtained in this way (12.7 ± 1.2, $r^2 = 0.6$) is not significantly different from the value obtained using Jungfraujoch as background site. Please note that the JFJ background

represents a clean tropospheric background with a footprint covering a large part of Europe. In the manuscript, we have stated that the R_{CO} value is almost insensitive to the choice of CO background. In both cases, the background values are derived using the REBS smoothing technique.

L371: The authors mention that cold conditions and mass transport from Eastern Europe are likely causes, yet no meteorological data is shown in this paper. Please consider adding a supplement with the key information that you have based this analysis on.

As mentioned above we have now included the model simulations for the air masses during each sampling period in the supplementary materials. We have also added the temperature data from the Beromünster tower at 212.5 m in Figure 2.

L380: The ratios do indeed differ significantly, but you need to establish why Beromünster-JFJ based RCO should be representative for Switzerland (only). See general comment #3

According to a thorough footprint analysis of the Beromünster tower by Oney et al., (2015) (Figure 12) the Beromünster tower foot print is restricted to Switzerland during winter and the R_{CO} values are independent of which background site used while in summer it includes larger areas from neighboring Germany and France. However, during winter/spring air-masses transported from Eastern Europe were reported (Oney et al 2017) and shown from the FLEXPART-COSMO simulations (Supplementary materials).

L407: Colder temperatures are mentioned the main cause, again. If the temperature is such a good predictor of DCO2ff a simple scatter plot should suffice to strengthen your argument.

The temperature record is now additionally included to in Fig. 2.

L425: Please consider adding visual aides to highlight the seasonal cycle in Figure 5. It seems not too clear in the printed version.

Figure 5a shows the all year time series of CO_{2bio} but the data seems noisy due to strong day-night variation in CO_{2bio}. However, clear signals are also present for e.g. negative spikes are frequent during summer implying net uptake. Adding a seasonally varying harmonic fit to show seasonal pattern will be misleading considering the strong variability in CO_{2bio} but averaging these values as shown in Figure 6 better illustrates the seasonal variations.

Table 1: An overall of 45 RCO values is reported, while the study is supposedly based on a 3-years long time series (L451) of bi-weekly samples (i.e. 78). Seven samples were accounted for due to the leakage problem reported in L167. What caused the other 26 to be missing here? Were they excluded, not samples, etc?

It is indeed an important point as mentioned by the reviewer that the lower sample amount need clarification. The study period in this manuscript is from 31 July 2013 until 3 December 2015, which is 2.3 years, now corrected in line 508.

During this period, we have collected one sample per month until October 2013 and the biweekly sampling started from December 2013. Hence we have collected 7 samples in 2013, 26 samples in 2014 and 24 samples in 2015 with a total of 57 samples, out of which 7 were excluded due to contamination. From the remaining 50 samples, 5 were excluded due to a strong mismatch among the triplicates in terms of CO_2 amount after sample extraction which is indicative of contamination. We have now clarified these points in lines 144 and

182.

No the dashed line in Figure 2c indicates the mean CO_{2ff}. The one in Fig. 2d is just y=0 ppm, to indicate the sign of the CO_2 signal and it is now changed into a solid line.

See comments above

Henne, S., Brunner, D., Oney, B., Leuenberger, M., Eugster, W., Bamberger, I., Meinhardt, F., Steinbacher, M., and Emmenegger, L.: Validation of the Swiss methane emission inventory by atmospheric observations and inverse modelling, Atmos Chem Phys, 16, 3683-3710, 2016.
Oney, B., Henne, S., Gruber, N., Leuenberger, M., Bamberger, I., Eugster, W., and Brunner, D.: The CarboCount CH sites: characterization of a dense greenhouse gas observation network, Atmos. Chem. Phys., 15, 11147-11164, 10.5194/acp-15-11147-2015, 2015.